Electronic Supplementary Material (ESI) for RSC Advances.

Supporting Information

N-Methylpyrrolidinium Hydrogen Tartrate (NMPHT): An Above-Room-Temperature Order–Disorder Molecular Switchable Dielectric Material

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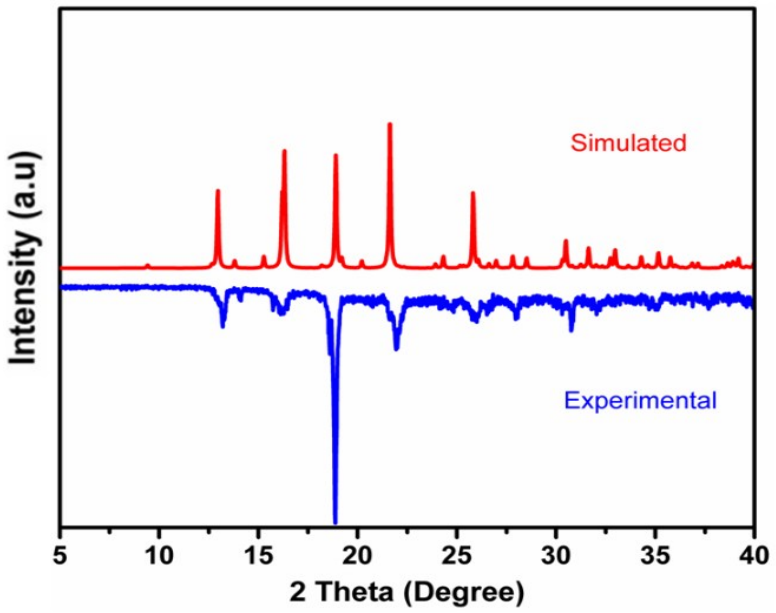

Figure S1. Experimental and simulated PXRD Patterns of **NMPHT**, verifying the phase purity.

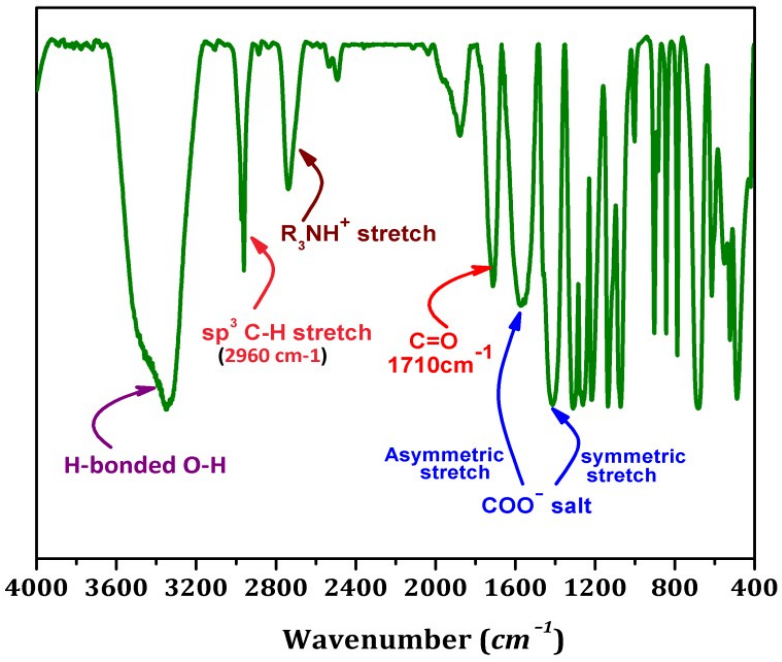

Figure S2: IR Spectrum of **NMPHT,** showing both the carboxylate and carboxylic acid groups.

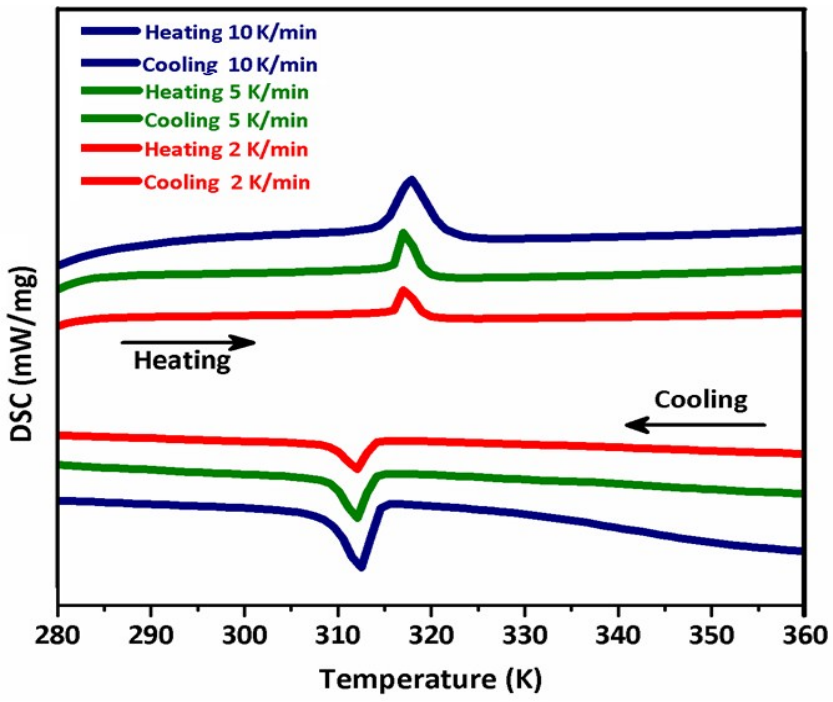

Figure S3. DSC curves of **NMPHT** at different scanning rates (2 K/min, 5 K/min, and 10 K/min).

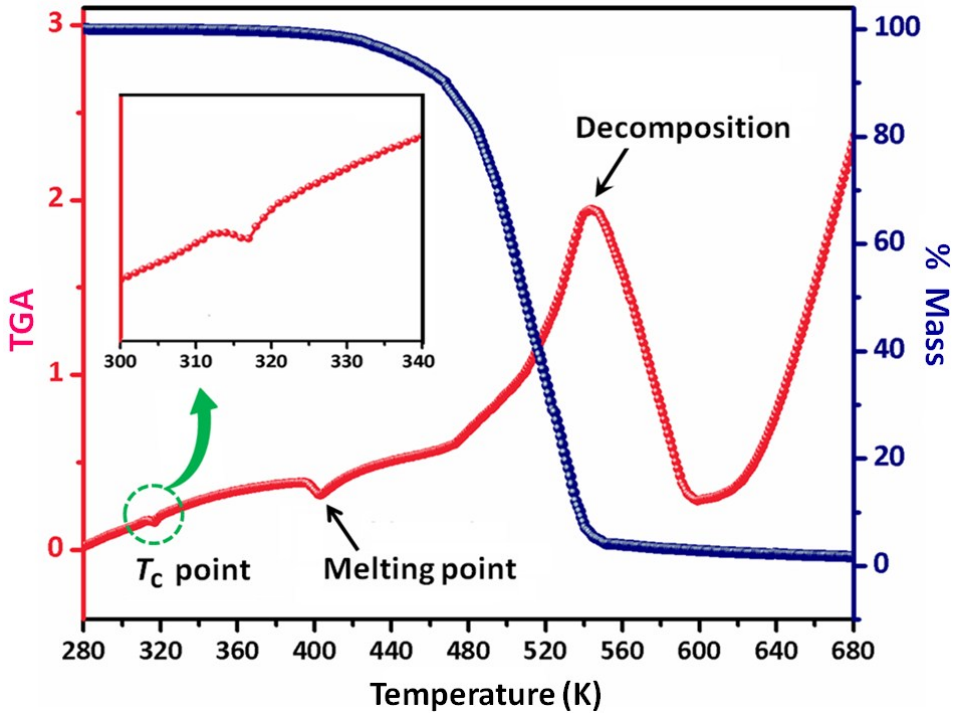

Figure S4. Differential thermal analysis (DTA) and Thermo-Gravimetric (TG) curves of **NMPHT**. The T_c point appears as a small protuberance, represented on the left side.

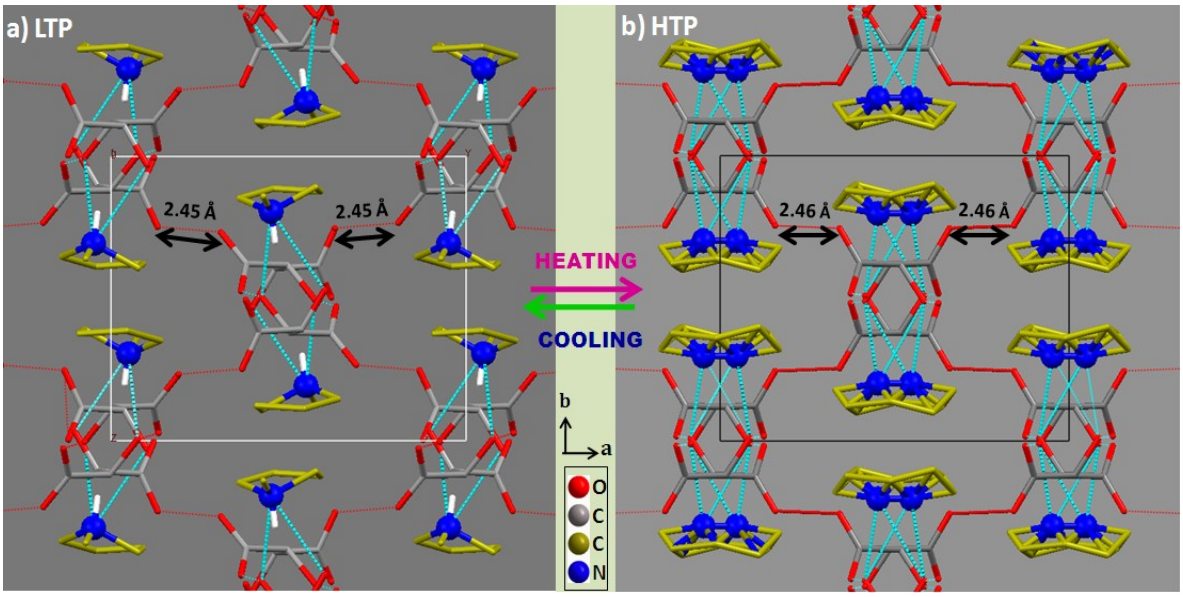

Figure S5: The packing views of **NMPHT** along the c-axis at (a) LTP and (b) HTP. Carbon-bonded H-atoms are omitted for clarity. The hydrogen bonds are represented as dashed lines.

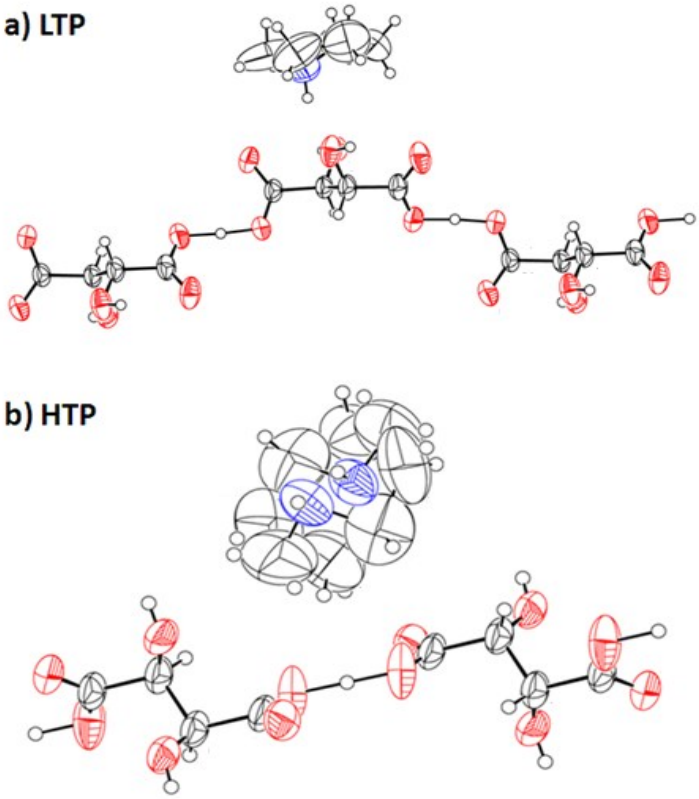

Figure S6. Thermal ellipsoidal view of **NMPHT** at (a) LTP and (b) at HTP.

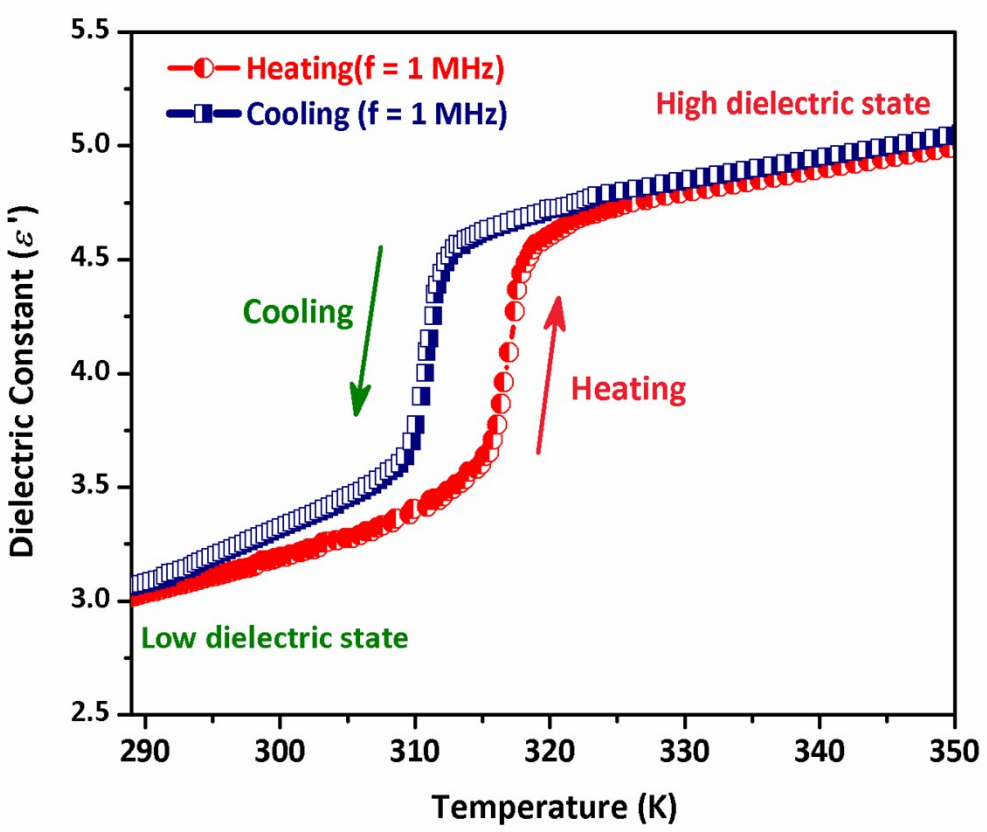

Figure S7. Temperature dependent cycle of dielectric constant of **NMPHT** at 1 MHz, showing the reversible dielectric switching between high and low dielectric states**.**

Table S1: The SCXRD data and structure refinement details of **NMPHT** at 330 and 260 K.

Temperature (*K*)	330*K*, Cu-*K*α	260*K*, Cu-*K*α
Sum formula	$C_9H_{17}N_1O_6$	$C_9H_{17}N_1O_6$
Moiety formula	C_4 H_5 O_6, C_5 H_{12} N	C_4 H_5 O_6, C_5 H_{12} N
Formula Weight	235.24	235.24
Crystal system	Monoclinic	Monoclinic
Space group	*C* 2/*c*	*P* 2_1/*n*
Cell parameters	*a*= 11.7014(5) Å	*a*= 11.3346(4) Å
	b= 8.7644(3) Å	*b*= 8.7794(3)Å
	c= 12.0457(3)Å	*c*= 12.0991 (4) Å
	α= 90°	α= 90°
	β= 107.298(4)°	β= 106.683(4)°
	γ= 90°	γ= 90°
Volume (Å^3)	1179.48(8)	1153.32(7)
Z	4	4
$D_{calcd.}$ (g/cm^3)	1.325	1.355
F(000)	504.0	504.0
Theta range (°)	3.8250 – 73.0240	4.6870 – 73.3100
Completeness	99.4%	99.6%
GOF	1.045	1.073
R_1 [on $F_\circ^2$ *I* > 2σ(*I*)]	0.0764(792)	0.0802(1699)
wR_2 [on $F_\circ^2$ *I* > 2σ(*I*)]	0.2587(1035)	0.2435(2034)

αR_1 = Σ||F_o| - |F_c| / Σ |F_o|, wR_2 = [Σ (|F_o|^2 - |F_c|^2) / Σ|F_o|^2]$^{1/2}$

Calculation of 'ΔS' and '*N*' for NMPHT:

According to Boltzmann equation:

$$\Delta S = nR\ln N$$

Where;

Δ*S* = entropy change,

R = the universal gas constant,

n = the number of guest molecules per mole (n =1, here), and

N = the number of possible orientations for the disordered system.

The *ΔS* and *N* values on the heating process of **NMPHT** are calculated as follows: [1]

$$\Delta S = \int_{T_1}^{T_2} \frac{Q}{T}\, dT$$

$$\Delta S \cong \frac{\Delta H}{T_c}$$

For 1 mole;

$$\Delta S = \frac{9.828\ J\ g^{-1} \times 235.24\ g\ mol^{-1}}{317.5\ K}$$ (where; 235.24 g mol^{-1} is the molecular mass of **NMPHT**)

$$\Delta S = \frac{2311.93\ J\ mol^{-1}}{317.5\ K}$$

$$\Delta S = 7.281\ J\ mol^{-1}K^{-1}$$

$$\Delta S = R\ln N$$

$$N = e^{(\frac{\Delta S}{R})}$$

$$N = e^{\left(\frac{7.281\ J\ mol^{-1}K^{-1}}{8.314\ J\ mol^{-1}K^{-1}}\right)}$$

$N = 2.40$

Reference:

1. Y.-Z. Tang, Z.-F. Gu, J.-B. Xiong, J.-X. Gao, Y. Liu, B. Wang, Y.-H. Tan and Q. Xu, *Chem. Mater*., 2016, **28**, 4476−4482.